Transport in (10,10) Carbon Nanotubes with Defects

M.P. ANANTRAM,[a] JIE HAN, AND T.R. GOVINDAN[b]

NASA Ames Research Center, Mail Stop T27-A, Moffett Field, California 94045-1000, USA

[b]*Applied Research Laboratory, P.O. Box 30, State College, Pennsylvania 16804-0030, USA*

ABSTRACT: In this paper, we numerically calculate the conductance characteristics of a uniform (10,10) carbon nanotube in the presence of an applied bias. It is shown that for long tubes, the conduction mechanism involves only the four nondegenerate sub-bands when the applied bias drops slowly across the tube. The focus of our work is to calculate the transmission coefficient and low bias conductance of carbon nanotubes with defects. Our calculations show that (1) a weak uniform disorder in the tube does not cause a significant change in the zero bias conductance and (2) substitutional defects which are strong scatterers, can cause a gap in the transmission coefficient versus energy at the middle of the band resulting in a significant change in the low bias conductance

INTRODUCTION

Carbon nanotubes (CNT) are hollow cylindrical surfaces of carbon atoms with the lattice structure of a rolled up graphite sheet. They can presently be fabricated as both multi-walled or single-walled tubes, with the multi-wall tubes typically containing concentric cylinders of single-wall tubes.[1] Experimentally, CNT have demonstrated use as STM tips[2] and room temperature electron field emitters.[3] The possibility of making wires and devices led to transport experiments on single-wall nanotubes and their ropes, which measured the low bias conductance as a function of temperature, gate voltage, and magnetic field,[4–6] and the I-V characteristics.[7] The experimental activity in manufacturing CNT coupled with the prospect of some useful applications has generated considerable interest in modeling various properties of these tubes. Calculations have shown that the Young's modulus is comparable to 1 TPa; tubes with different diameters and chirality can have properties varying from that of semiconductors to metals; the low bias conductance can be as large as $4e^2/h$ (molecular wires) and junctions between semiconducting and metallic tubes can be formed.

Defect-free zig-zag and armchair tubes have two propagating modes at the center of band (Fermi energy for infinitely long tubes) with a maximum conductance of $4e^2/h$. To measure a low bias conductance that is comparable to $4e^2/h$ requires both good contacts to the large electron reservoirs where voltage is applied and absence of defects. Both factors lead to reflection of electrons in the CNT causing a decrease in conductance. This paper calculates the I-V characteristics of a uniform CNT in

[a]Address for correspondence: anant@nas.nasa.gov

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the presence of an applied bias and examines the influence of defects on the transmission coefficient and low bias conductance. The two types of defects we have considered are the *weak uniform disorder* and *substitutional defects*.

MODEL

The electronic properties are modeled using a single π electron per spin per atom in a sp^2 bonded network.[1,8,9] The corresponding single particle Hamiltonian is,

$$H = \sum_i \varepsilon_i c_i^\dagger c_i + \sum_{i,j} t_{ij} c_i^\dagger c_j(i)\,, \tag{1}$$

where, the summation is over all lattice sites of the tube. $\{c_i^\dagger, c_i\}$ represent the creation and annihilation operators at site i and $j(i)$ represents the three nearest neighbors of a carbon atom in the uniform tube. ε_i is the on-site potential, which was chosen to be zero for an uniform tube and the hopping parameter between neighboring carbon atoms t_{ij} was assumed to be −3.1 eV.[8]

The current, across the device was determined by assuming the contacts to be in local equilibrium with all unique chemical potential assigned to them. We calculate the current in the phase coherent limit, where the effect of phonons and dephasing is neglected. In this limit, the current (I) is related to the Fermi-Dirac factors in the contacts ($f_{\text{source}}(E)$ and $f_{\text{drain}}(E)$) and the transmission coefficient $T(E)$ between the contacts, by the Landaue-Buttiker formula,[11]

$$I = \int dE\, T(E)(f_{\text{source}}(E) - f_{\text{drain}}(E)), \tag{2}$$

where E is the energy coordinate. The temperature enters the calculations only through the Fermi factors of the contacts. That is, we assume that the lattice is at zero temperature and the electrons are not. In computing the transmission coefficient, the spatial variation of the electrostatic potential and the modeling of the connection between the CNT and contacts must be considered. Calculating the contribution of the electrostatic potential through the wire requires a self-consistent solution of the equation for charge density and Poisson's equation. With the objective to obtain broad features of the I-V characteristics, we have calculated the current for different assumed forms of the applied potential. The electrostatic potential in the tube was assumed to have the form,

$$V(x) = V_{app}/2 + Ae^{+x/L_{sc}} + Be^{-x/L_{sc}} \tag{3}$$

where

$$A = -\frac{1}{2}\frac{1+e^{-L/L_{sc}}}{e^{L/L_{sc}} - e^{-L/L_{sc}}} \text{ and } B = +\frac{1}{2}\frac{1+e^{+L/L_{sc}}}{e^{+L/L_{sc}} - e^{-L/L_{sc}}}$$

where L is the length of the wire and L_{sc} is a parameter that can be varied to yield different forms for the potential drop. For values of $L_{sc} > L$, $V(x)$ varies linearly

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across the wire and for values of $L_{sc} < L$, the potential drops abruptly close to the ends of the tube, with the V potential in the bulk of the wire being $V_{app}/2$. Assuming the presence of an adiabatic connection[12] between the contacts and the device, the transmission coefficient was calculated using semi-infinite CNT leads as boundary conditions.

In the case of uniformly distributed weak disorder, small and random variations in the location of the atoms occur due to their interaction with a nonuniform surrounding. As a result, both the on-site potential and hopping parameters acquire a random component. Substitutional defects, on the other hand, correspond to lattice locations onto which electrons cannot hop easily. Our calculations are based on modeling these defects by exclusively varying the on-site potential. The modified on-site potential was $\varepsilon_i^0 + \delta\varepsilon_i$. In a uniformly distributed weak disorder, $\delta\varepsilon_i$ was chosen randomly from the range $\pm\varepsilon_{\text{random}}$ at every lattice point. The degree of disorder is proportional to $\varepsilon_{\text{random}}$. $\delta\varepsilon_i$ was set to a large number at lattice sites chosen randomly in the case of the substitutional defect.

RESULTS

Defect-Free Wire

For a uniform (10, 10) arm chair tube having twenty atoms per unit cell, FIGURE 1 shows the transmission coefficient and k vector versus energy along the tube axis.[1] Of the forty quasi one-dimensional bands, eighteen are twofold degenerate (*solid lines*) and four are nondegenerate (*dashed lines*). The transmission coefficient increases (decreases) every time a sub-band opens (closes) The change in transmission coefficient by units of two corresponds to the opening and closing of the twofold degenerate sub-bands. Also, note that while the nondegenerate sub-bands meet at $E =$ 0 eV, the degenerate bands have a large band gap.

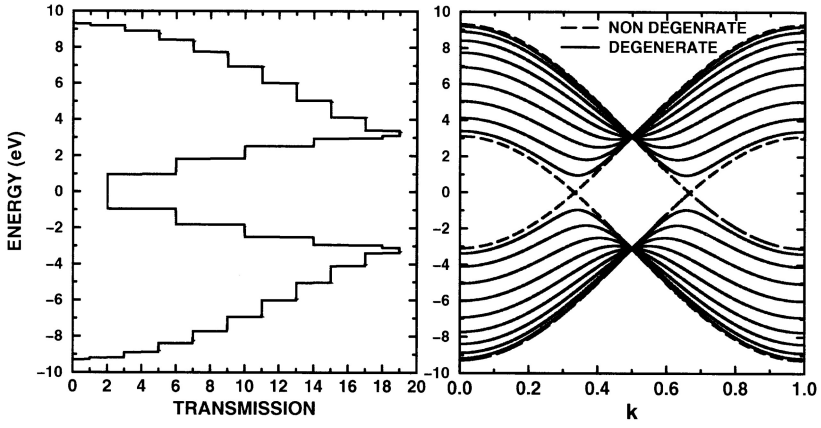


FIGURE 1. **(a)** Transmission and **(b)** k vector versus energy of a (10,10) tube. The transmission increases/decreases when a sub-band in **(b)** opens/closes, The *dashed* and *solid lines* are non-degenerate and degenerate sub-bands respectively.

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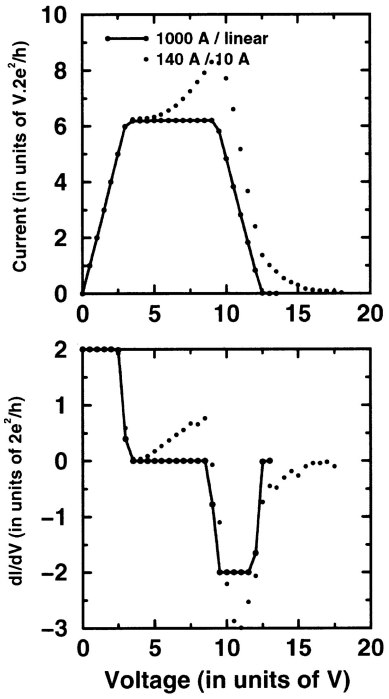


FIGURE 2. The current-versus-voltage curve for a tube of length 1000 Å and L_{sc} = 2000 Å, which corresponds to a linear drop in the potential across the tube (*solid line*). The dots correspond to a wire of 140 Å and L_{sc} = 10 Å. The lower curve is *dI/dV*.

In the presence of a finite applied bias, we have calculated the current versus voltage for different values of length (L) and L_{sc}. For a 1000 Å wire with L_{sc} = 2000 Å (which corresponds to a linear drop in the potential across the tube), the current-versus-voltage characteristics are shown in FIGURE 2, which can be interpreted as follows. In the presence of a slowly varying potential across the tube the degenerate bands (*solid lines* in FIG. 1b) do not contribute to transport because of the finite band gap between the conduction and valence bands. Further, for a slow variation of the applied potential, band-to-band tunneling is small. Electrons incident from the source contact in the two positive velocity states (source to the left and drain to the right) of the nondegenerate bands gain velocity and k vector as they move to the right and are injected into the drain for applied voltages less than 3.1V. This holds good for electrons in the energy regime of −3.1 eV to 0, which is taken to be the Fermi energy in the source end. The transmission of two states per energy from these quasi one-dimensional bands leads to the conductance of $4e^2/h$ (spin degeneracy included) in a straightforward manner from Eq. (2). In contrast, for voltages larger than 3.1V, the k vector increases to values where Bragg reflection of incident positive velocity states at some energies occurs (for example, close to the Fermi energy of the source). However, this happens in such a way that the current remains fixed until about 9.3V, when the band edge in the drain end of the tube and the Fermi energy of the source are comparable. Further increase in the applied voltage decreases the current. The overall feature of the decrease in current with increase in voltage can be understood as follows. Near the source end, the π electron band structure of the CNT extends

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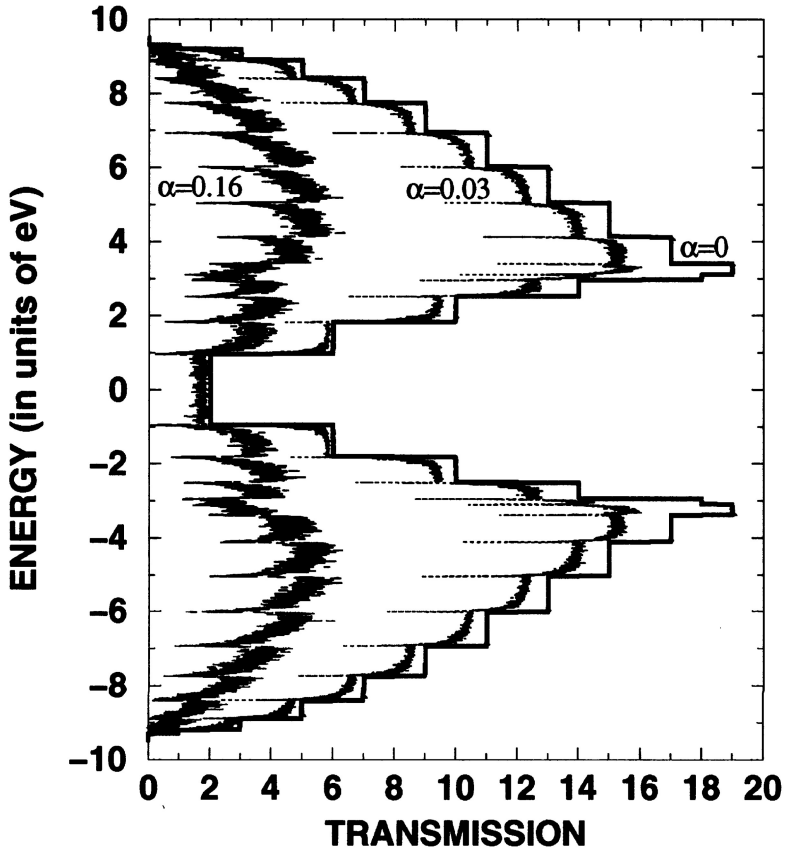


FIGURE 3. The transmission coefficient versus energy in the presence of disorder for a 1000 Å long tube. The disorder increases with increase in $\alpha = \varepsilon_{random}/3.1$ (see text).

from $-9.3eV + eV_{app}$ to $+9.3eV + eV_{app}$ and near the drain end, the band structure extends from $-9.3eV$ to $+9.3eV$. For current flow at an energy, the Fermi factor at this energy at the source and drain should be different and also an incident electron from the source (drain) at this energy should be able to scatter into a state at the drain (source) end at the same energy. The overlap between the bands at the source and drain ends decrease with increasing applied voltage, and for a large enough applied voltage the overlap becomes so small that the current begins to decrease before becoming zero. Note that this is an idealized picture that has neglected the effects of phonons and the other bands that are present in a more sophisticated band structure calculation.

The semiclassical discussion in the previous paragraph is not fully correct when the voltage drops across a short tube or if the screening length is very small because for large values of electric field in the tube, tunneling events between sub-bands,

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which were negligible at smaller fields, begin to contribute to the current.[13] An example of this is shown using a tube of length 140 Å and L_{sc} = 10 Å (FIG. 2). The deviation in the conductance at the larger applied voltages was consistent with increasing significance of tunneling events at greater electric fields.

Disordered Wires

FIGURE 3 illustrates the case of weak disorder distributed randomly in a tube of length 1000 Å. It is a plot of the transmission coefficient versus energy for various strengths of disorder. Our main observations are: (1) this type of disorder does not change the transmission coefficient significantly at energies near the center of the band; (2) the transmission coefficient versus energy develops sharp resonances; and (3) the transmission coefficient changes by large amounts near the opening and closing of sub-bands.

Weak disorder in the on-site potential introduces randomly distributed quasi-bound states along the length of the tube.[13] Tunneling across the tube via these quasi-bound states causes the resonances in FIG. 3. By increasing the degree of disorder,

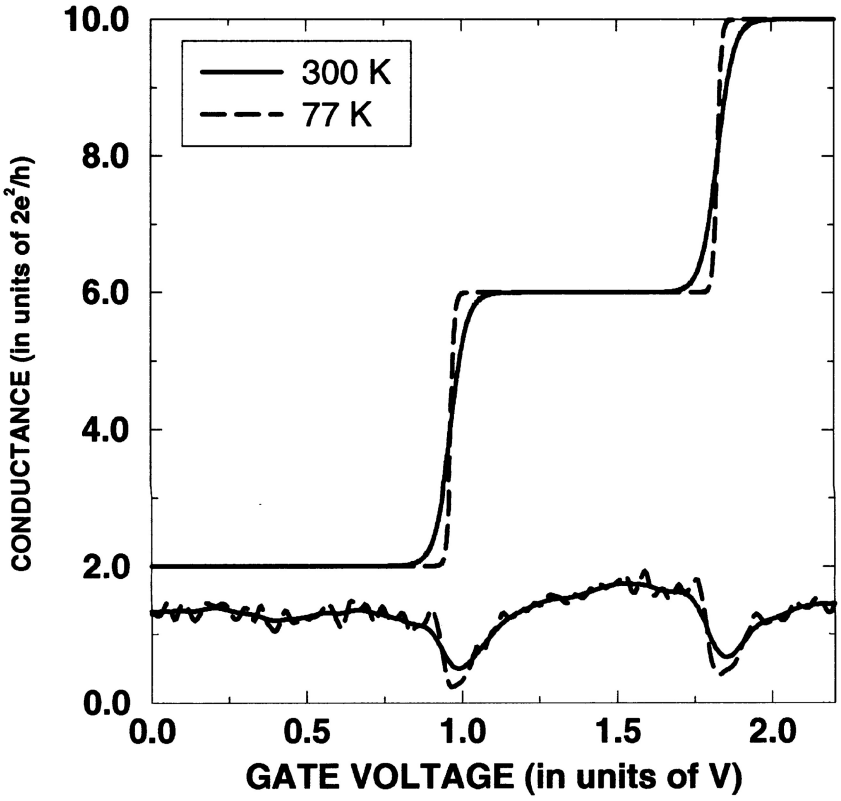


FIGURE 4. Small bias conductance versus gate voltage for the case shown in FIGURE 3. The conductance does not change significantly close to zero gate voltage.

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the quasi one-dimensional states localize more strongly in position, leading to an overall decrease in the transmission coefficient. The decrease in the transmission coefficients at sub-band openings and closings in FIG. 3 was in accordance with the explanation that an infinitely long structure with a single sub-band localizes at the band edges, where the velocity is small.[13] A particle incident in one of the nondegenerate sub-bands (*dashed lines* of FIG. 2) at energies close to the second sub-band opening (~0.9 eV) scatters to states at the band edge of the second sub-band. This leads to reflection of the incident state as these low velocity states in the second sub-band are localized. As a result, there is a (111) in the transmission coefficient. The low bias conductance versus the gate voltage calculated using Eq. (2) indicated that the CNT behaves as a good wire, in the presence of weak uniform disorder (FIG. 4) when the Fermi energy was close to the band center. A much larger decrease in the conductance was realized when the Fermi energy was close to a sub-band opening or closing.

In the presence of a single substitutional defect, it was observed by Chico *et al.*[9] that the transmission coefficient decreased at all energies with a dip in the center of the band. The transmission coefficient versus energy characteristics was computed in the presence of many such defects to see if the conductance of $4e^2/h$ was affected significantly. Results are presented for a tube of 140 Å in length with ten defects sprinkled randomly along the tube. The most prominent feature in the transmission versus energy plot (FIG. 5) is that a gap opens up in the middle of the band. A few narrow resonances traverse the entire tube. Our simulations show that increasing the

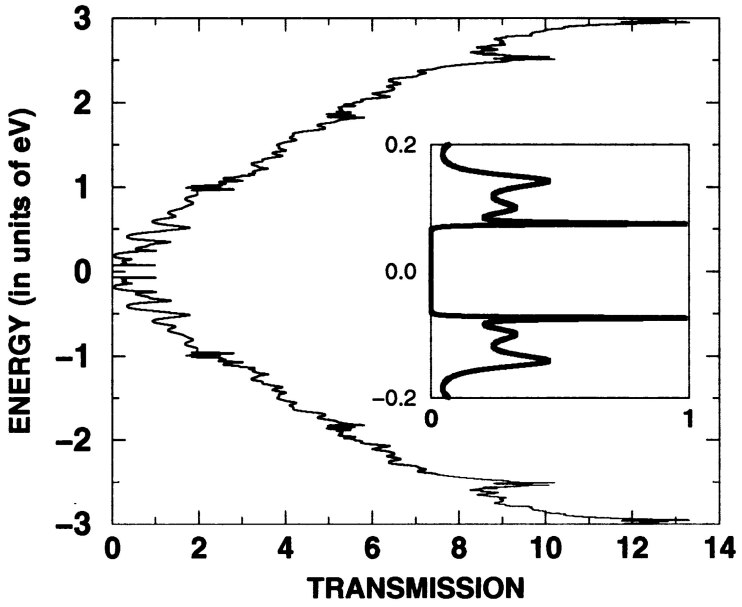


FIGURE 5. Transmission versus energy for a 140 Å long tube in the presence of substitutional defects. A gap opens in the transmission at the middle of the band (*inset*).

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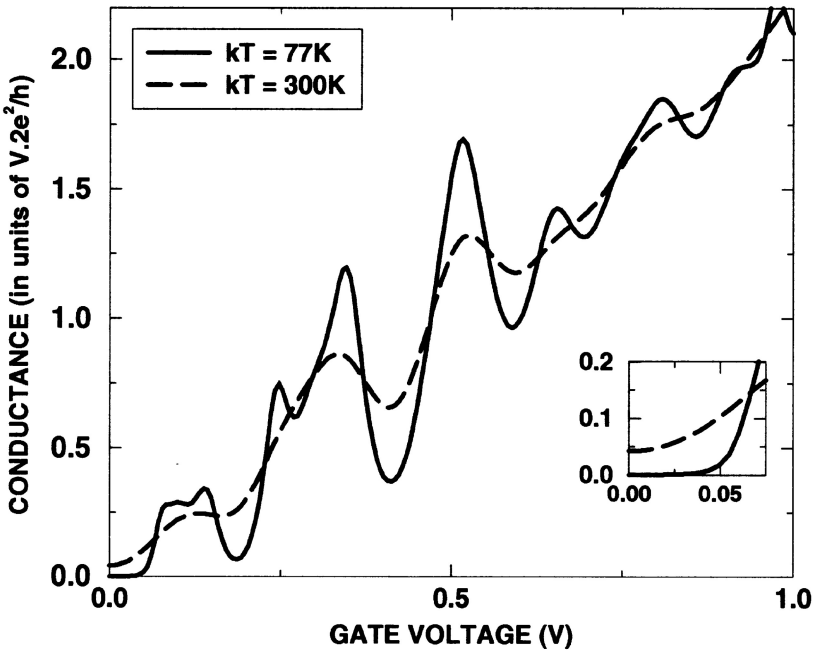


FIGURE 6. Small bias conductance versus gate voltage corresponding to the case in FIGURE 5. Note the significant drop in conductance close to zero gate voltage and the corresponding temperature dependence (*inset*).

number of defects removed these resonances in the transmission gap and further broadened it. The second feature was the stepwise increase in the transmission coefficient (FIG. 1) diminished leading to a comparatively linear behavior. Thirdly, similar to the previous case of disorder discussed, the transmission coefficient developed sharp resonances at energies corresponding to opening and closing of sub-bands. The conductance was calculated from Eq. (2). While the conductance was almost temperature independent in the defect-free case, in the presence of these defects, the conductance was strongly temperature dependent (FIG. 6). The small bias conductance close to zero bias was about forty times smaller than the defect-free value of $4e^2/h$ at a temperature of 300 K. Simulations were performed with different configurations of the defects and results obtained were qualitatively similar.

SUMMARY

The conductance properties of uniform tubes with and without defects were numerically calculated. Typical lengths of the CNT in these calculations were 140 Å and 1000 Å. In the defect-free case, the current-versus-voltage characteristics were computed for two different forms of the potential drop across the tube. We observed

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